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Aerosol pH and chemical regimes of sulfate formation in aerosol water during winter haze in the North China Plain

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Understanding the mechanism of haze formation is crucial for the development of deliberate pollution control strategies. Multiphase chemical reactions in aerosol water have been suggested as an important source of particulate sulfate during severe haze (Cheng et al., 2016; Wang et al., 2016). While the key role of aerosol water has been commonly accepted, the relative importance of different oxidation pathways in the aqueous phase is still under debate, mainly due to questions about aerosol pH. To investigate the spatio-temporal variability of aerosol pH and sulfate formation during winter in the North China Plain (NCP), we have developed a new aerosol water chemistry module (AWAC) for the WRF-Chem model (Weather Research and Forecasting model coupled with Chemistry). Using the WRF-Chem-AWAC model, we performed a comprehensive survey of the atmospheric conditions characteristic for wintertime in the NCP, focusing on January 2013. We find that aerosol pH exhibited a strong vertical gradient and distinct diurnal cycle, which was closely associated with the spatio-temporal variation in the relative abundance of acidic and alkaline fine particle components and their gaseous counterparts. Over Beijing, the average aerosol pH at the surface layer was ~5.4 and remained nearly constant around ~5 up to ~2 km above ground level; further aloft, the acidity rapidly increased to pH ~0 at ~3 km. The pattern of aerosol acidity increase with altitude persisted over the NCP, while the specific levels and gradients of pH varied between different regions. In the region north of ~41°N, the mean pH values at surface level were typically >6 and the main pathway of sulfate formation in aerosol water was S(IV) oxidation by ozone. South of ~41°N, the mean pH values at surface level were typically in the range of 4.4 to 5.7, and different chemical regimes and reaction pathways of sulfate formation prevailed in four different regions, depending on reactant concentrations and atmospheric conditions. The NO₂ reaction pathway prevailed in the megacity region of Beijing and the large area of Hebei Province to the south and west of Beijing, as well as part of Shandong Province. The transition metal ion (TMI) pathway dominated in the inland region to the west and the coastal regions to the east of Beijing, and the H₂O₂ pathway dominated in the region extending further south (Shandong and Henan Provinces). In all of these regions, the O₃ and TMI pathways in aerosol water as well as the gas-particle partitioning of H₂SO₄ vapor became more important with increasing altitude. Although pH is sensitive to the abundance of NH₃ and crustal particles, we

show that the rapid production of sulfate in the NCP can be maintained over a wide range of aerosol acidity (e.g., pH = 4.2-5.7) with transitions from TMI pathway dominated to NO₂/O₃ pathway dominated regimes.